SMX 3161.2 (98-13DIV2)
PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Application of Shenheng Guan et al. Art Unit 1743
Serial No. 10/071,546
Filed February 8, 2002
Confirmation No. 9873
For APPARATUS FOR SCREENING CATALYSTS IN A PARALLEL FIXED-BED REACTOR
Examiner Dwayne K. Handy

DECLARATION OF PRIOR INVENTION UNDER 37 CFR § 1.131

I, Shenheng Guan, declare as follows:

1. I am currently an associate adjunct professor in the Department of Pharmaceutical Chemistry at the University of California, San Francisco.

2. From October, 1996 to January, 2002, I was employed as a Distinguished Scientist at Symyx Technologies, Inc. (Santa Clara, California), the assignee of the above-referenced U.S. patent application, Serial No. 10/071,546.

3. I am named as a joint inventor in the above-referenced patent application and I am the sole inventor of some of the claimed subject matter, including the invention defined in independent claims 61-63 and dependent claims 70 and 80-87 depending therefrom as currently amended and presented in Amendment C.

4. I conceived of and, with the assistance of other Symyx employees, reduced to practice in the United States the subject matter of the invention as defined in currently amended independent claims 61-63 and dependent claims 70 and 80-87 depending therefrom before February 19, 1998, the facts being set forth below.

5. The invention described and claimed in the subject patent application was developed as part of a materials research program that I participated in while employed at Symyx Technologies, Inc.

6. Exhibit A, attached to this Declaration, is a true and correct copy of an internal research report I prepared prior to February 19, 1998 in connection with the above-mentioned research program as part of my duties at Symyx Technologies, Inc. The report describes the conception and development of a parallel fixed-bed reactor for screening catalysts and evinces my conception of the subject matter of currently amended independent claims 61-63 and dependent claims 70 and 80-87 depending therefrom and reduction to practice of an operational "six-channel prototype reactor" prior to February 19, 1998.

7. Exhibit B, attached to this Declaration, is a true and accurate copy of laboratory notebook pages I prepared prior to February 19, 1998 in accordance with established practices while employed at Symyx Technology, Inc. These notebook pages were prepared and include data recorded in connection with screening samples of molybdenum-vanadium-niobium oxide (Mo-V-Nb-O) catalysts in the gas-phase dehydrogenation of ethane to ethylene using the above-referenced parallel fixed-bed reactor. In particular, on pages 86-89 of the notebook, I carried out a number of experimental runs using the six-channel parallel fixed-bed reactor and catalysts of varying composition. In these experiments, reaction vessels 1-3 contained a sample of a particular catalyst (designated in the notebook as CX 1023 through CX 1029) and reaction vessels 4-6 contained a sample of a catalyst of different composition (designated in the notebook as CX 2023 through CX 2029). The composition of the gaseous effluents exiting the reaction vessels were measured using two gas chromatographs (GCs) containing three identical GC channels. The reactions were carried out at approximately atmospheric pressure and the temperature was controlled at about 300°C. The actual temperature measured in the reaction vessels is recorded in the notebook pages along with the GC peak area readings for CO, CO_2, and C_2H_4 measured in the respective gaseous effluents exiting the six-channel parallel fixed-bed reactor during dehydrogenation of ethane to ethylene. The data recorded in these notebook pages was used to determine the conversion and selectivity obtained using the different catalysts.

8. The data recorded in the notebook pages of Exhibit B are in part the basis for my statement in Exhibit A that "Currently, the six-channel prototype reactor is operational and producing reliable data."

9. All work referred to herein was carried out in the United States.

10. All statements that I made herein of my own knowledge are true and all statements made on information and belief are believed to be true. I acknowledge that willfully making false statements is punishable by fine, imprisonment, or both, under 18 U.S.C. § 1001 and that such willful false statements may jeopardize the validity of any patent issuing from this application.

Shenheng Guan

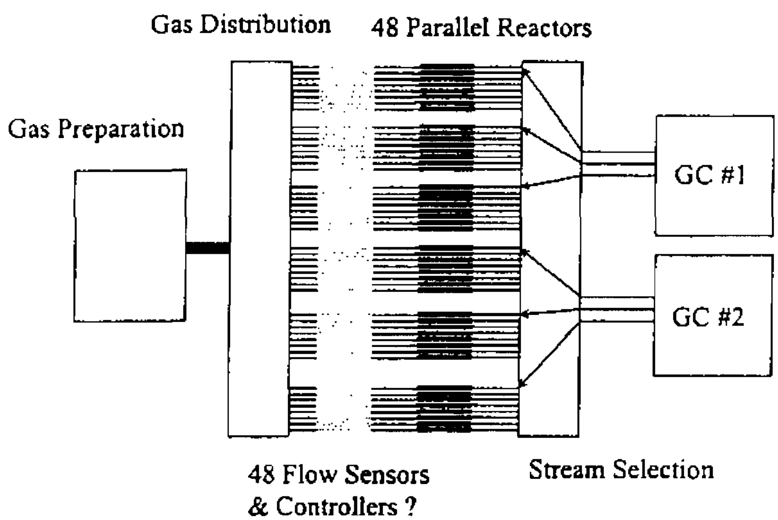
9/28/06
Date

Symyx Parallel Fixed Bed Reactor

Shenheng Guan

The Design philosophy of the parallel fixed bed reactor is to scale down from conventional laboratory reactors and to operate many reactors simultaneously. By choosing catalyst loading to be two orders of magnitude lower than that in conventional reactors, many catalysts can be synthesized quickly. Since many important parameters, such as space velocity, contact time, and power forms of catalyst, can be preserved, data from the reactor can often be compared with that from conventional methods directly. Final version of the parallel reactor contains 48 individual reactors.

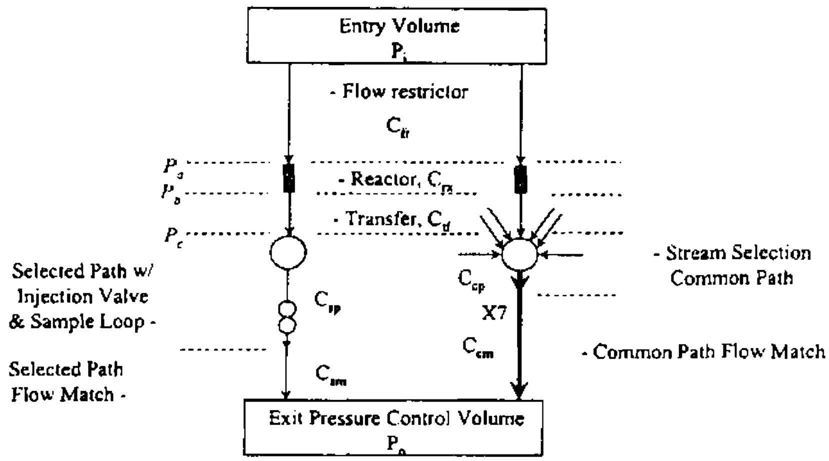
Parallel Reactor Flow Geometry



Design of the parallel fixed bed reactor offers several advantages over single bed reactors. It allows creation of nearly identical reaction conditions for all the individual reactors and allows one to carry out catalyst activation and catalytic reactions simultaneously. With a careful design, reaction conditions for the individual reactors can be controlled – creating opportunities for combinatory studies of reaction conditions for the same catalyst. Six identical gas chromatography (GC) channels analyze products simultaneously. Since 48 reactors need to be analyzed, sophisticate gas distribution and stream selection valve control methods are introduced. By use of GC for detection, almost all possible reaction systems can be investigated. The parallelism of the reactor allows analysis process to be pipelined, therefore, characterization time can be minimized.

The key design concept of the reactor is to use flow restriction up-stream from individual reactors. By supplying a common inlet pressure and a common outlet pressure, the flow rate of the individual reactor is determined mainly by conductance of the flow restriction.

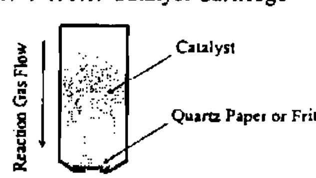
Parallel Reactor Flow Matching



If the flow restriction has conductance orders of magnitude smaller than that of the rest flow path and all reactors have the same flow restriction, the variation of conductance among reactors (for example, by catalyst packing) will not cause large variation in flow rate. Mass flow controllers premix reaction gas that is then supplied to all the reactors through a bundle of long and narrow bore capillaries (as flow restrictors). Flow measurement has been done on the six-channel prototype reactor. A flow variation of less than 2% was observed.

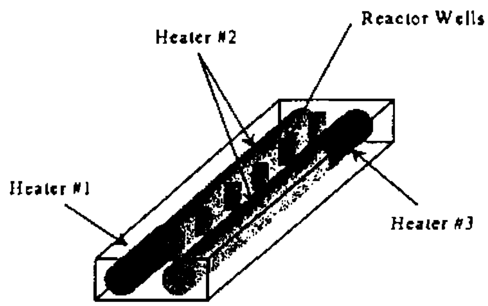
The prototype reactor block was constructed with stainless steel. Catalysts are contained in cartridges made of quartz or stainless steel.

Parallel Reactor Catalyst Cartridge



Quartz paper is used as frit to prevent power from escaping from the cartridge. The reaction block can be opened on the top to expose all the catalyst cartridges, allowing easy sample change. A three heating zone method is developed to achieve uniform temperature distribution among reactor wells.

Temperature Control Method in Parallel Reactor



The three zones are independently controlled with PID loops. A temperature spatial variation below 4°C at 350°C has been realized. The additional benefit from such method is a linear temperature gradient can be easily generated. A 10°C difference between adjacent reactors was obtained. Currently, the six-channel prototype reactor is operational and producing reliable data.

EXHIBIT B

Micro Fiched
On________

NOTEBOOK NO. L (23)

ISSUED TO SHENHENG GUAN

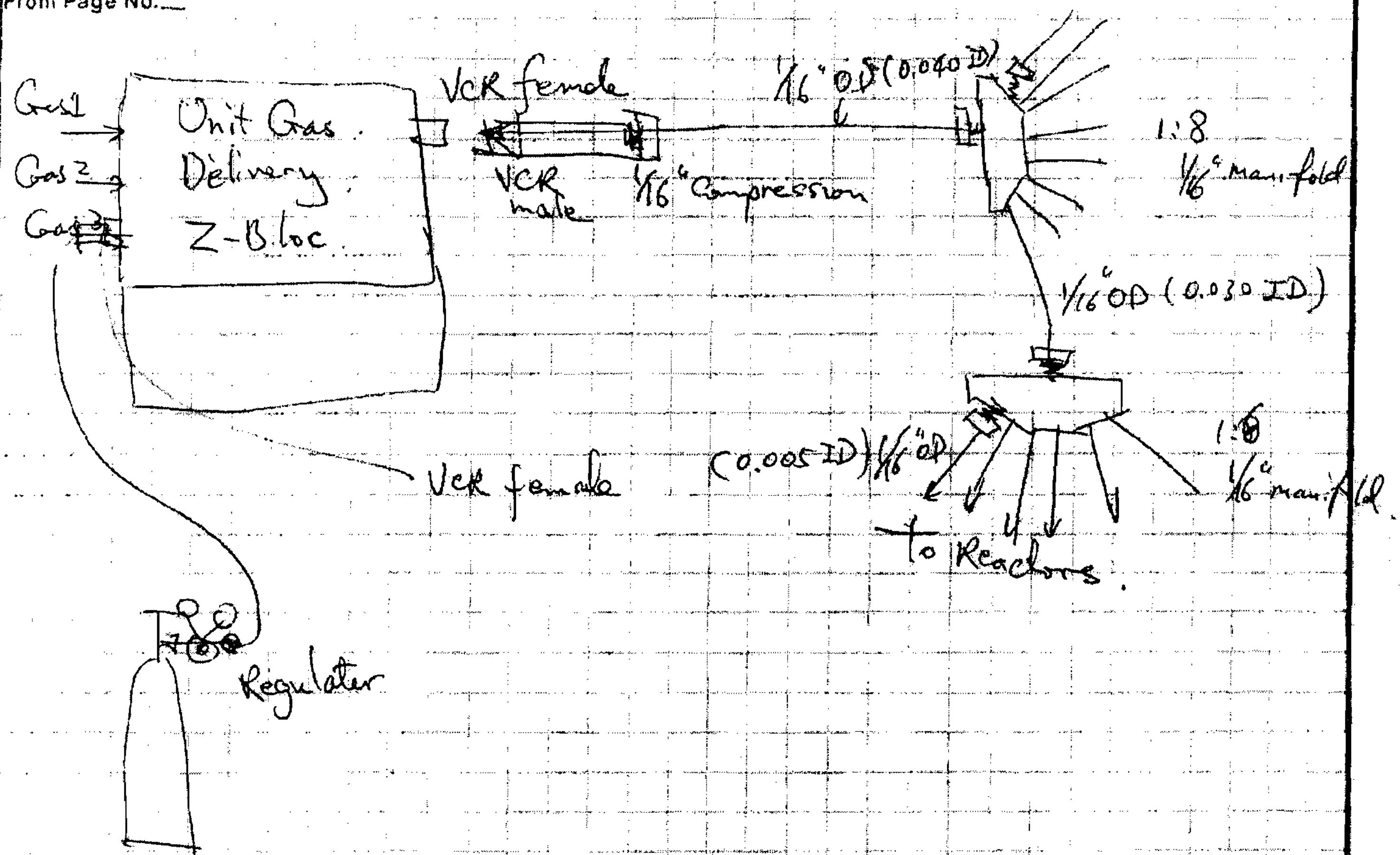
ON 10/14 **19** 96

DEPARTMENT Analytical Technology

RETURNED________ **19**____

Project No.
Book No.
TITLE Pipe Connection for Parallel Reactor

From Page No.

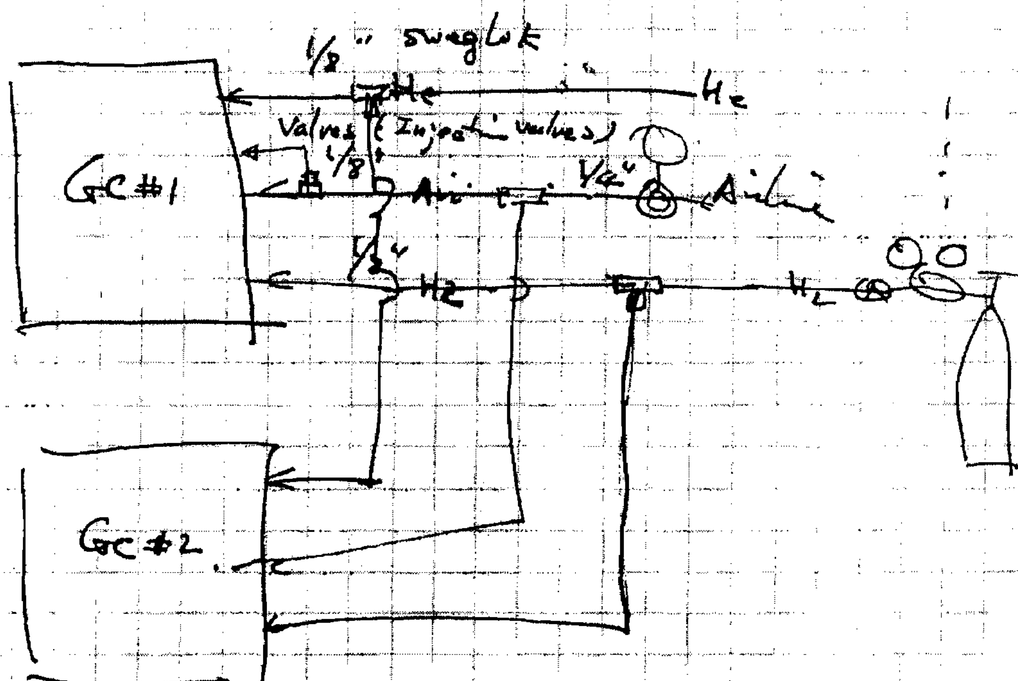


Regulater :	x 3	
1/4" NPT to 1/4" Compression	x 3	
5–10 m 1/4" OD line	x 3	
1/4" Compression to 1/4" VCR male	x 3	SS-4-VCR-6-400
VCR male to 1/16" Compression	X 1	SS-4-VCR-P
		CU-4-VCR-2
1/16" to 1/4" Reducer		SS-~~400~~-R-4

To Page No.

Witnessed & Understood by me,	Date	Invented by	Date
RABorade		[signature]	
		Recorded by SHENHENG GUAN	

From Page No. 84



To Page No.

Witnessed & Understood by me,	Date	Invented by	Date
RBBonde		Recorded by SHENHENG GUAN	

Project No.______
Book No.______ TITLE Fourth Milestone on Para Reactor

From Page No. 85

Flow : ch1 37.5% : 3.75 sccm 14.4% O_2 in N_2
ch2 5.0% : 2.50 sccm Ethane

P1 = 8 psig
P2 = 7 psig
P3 = 0 psig start 9:30 am

Temperature :

1	2	3	4	5	6	7
262.1	280.8	281.1	279.3	274.2	262.0	

File CX1023 Bad
CX2023 Good

	CO	CO_2	C_2H_6	Temp.
1	0.0889	0.0718	0.1408	262.1
2	0.0850	0.0870	0.1691	280.8
3	0.0856	0.0759	0.1747	281.1
4	0.344	0.1747	2.9458	279.3
5	0.2394	0.1713	3.080	274.2
6	0.1427	0.0992	2.232	262.0

To Page No.____

Witnessed & Understood by me, RMBorale | Date | Invented by [signature] | Date
Recorded by SHANHENG

From Page No. 86

Filue CX1024 Bad
CX2024 Good

	CO	CO_2	C_2H_4	Temp
1	0.0677	0.0638	0.1563	265.1
2	0.0814	0.0718	0.1844	280.5
3	0.0847	0.0655	0.1924	281.2
4	0.2727	0.2090	3.1700	280.0
5	0.2703	0.2057	3.325	276.1
6	0.1685	0.1257	2.513	265.2

Temp 280 → 300 CX1025
CX1025

1	0.1126	0.0939	0.2689	284.1
2	0.1331	0.1051	0.3150	300.9
3	0.1369	0.0967	0.3295	301.3
4	0.6164	0.5112	4.9116	300.0
5	0.6308	0.4812	5.1608	294.8
6	0.4157	0.3202	4.0752	284.1

To Page No. ____

Witnessed & Understood by me, RBMonde | Date | Invented by [signature] | Recorded by SHENHENG | Date

Project No.
Book No.
TITLE

From Page No. 87

CX1026
CX2026

	Temp.
1	285.1
2	301.0
3	301.3
4	300.0
5	293.7
6	286.5

Cx1027
Cx2027

	CO	CO_2	C_2H_4	
1	0.0662	0.1093	0.2717	285.5
2	0.0812	0.1167	0.3079	300.9
3	0.0924	0.1056	0.3254	301.2
4	0.6102	0.5378	4.9507	300.0
5	0.6530	0.4922	5.250	295.8
6	0.4612	0.3910	4.3359	287.2

To Page No.

Witnessed & Understood by me, RBBonde
Date
Invented by [signature]
Recorded by SHEN HENG
Date

TITLE ______ Project No. ______ Book No. ______

From Page No. 88

CX 2028
CX2028

1:22 pm 300°C → 350°C

	CO	CO_2	C_2H_4	Temp
1	0.2996	0.4040	0.8832	333.9
2	0.3587	0.4586	1.0216	350.9
3	0.3961	0.4097	1.0775	351.5 ~~333.~~
4	2.4481	1.9014	9.3465	350.1
5	2.7859	1.8087	10.1314	345.1
6	2.244	1.635	9.0623	334.8

CX1029
CX1029

300°C → 350°C

1	0.1151	0.1257	0.3904	300.
2				
3				
4				
5				
6	0.7949	0.5813	5.7231	300.3

To Page No. ______

Witnessed & Understood by me, RABonde

Date

Invented by [signature]

Recorded by JHEN HENG GOAN

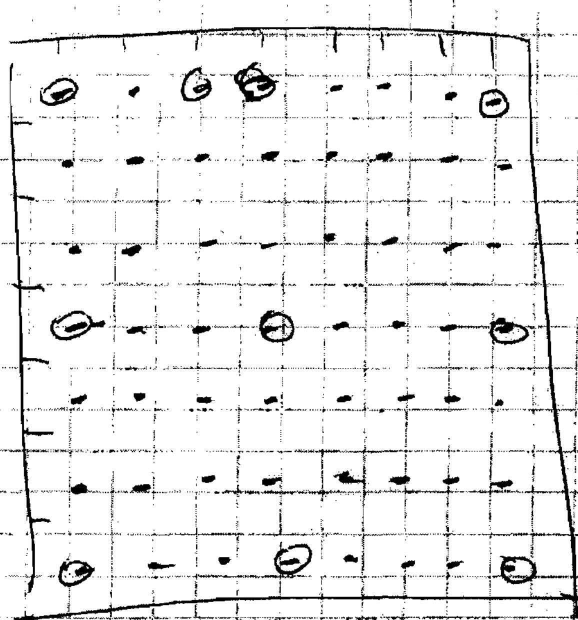
Date

Project No.____

Book No.____ TITLE ____

From Page No.__

AB Reactor thermocouple distribution



To Page No.____

Witnessed & Understood by me, RBBosade

Date

Invented by [signature]

Recorded by SHENHENG GUAN

Date

From Page No. ___

Xiaoping's Cat.

CX1001
CX2001

		Rx Temp	Catalyst preparation Temp (Treated)	
not compressed	1	282.7	400	4h
	2	301.0	400	8h
	3	301.2	400	12h
compressed	4	300.0	400	4h
	5	295.2	400	8h
	6	284.7	400	12h.

Flow changed to .

Ch I 75.0% 7.5 sccm 14.9% O_2 in N_2
Ch 2 . 10.0% 5.0 sccm Ethane

P1 : 1.2 psig
P2 : 12 psig
P3 : 0 —

CX1002
CX2002

To Page No. ___

Witnessed & Understood by me,	Date	Invented by	Date
RABonale		[signature]	
		Recorded by SHENHENG GUAN	

Project No.____
Book No.____ TITLE Temperature Measurement on PR.

From Page No.___

Top Blocknot Bolted Down

Temperature Setting 200.0°C @ Ch.4 for 30 mins

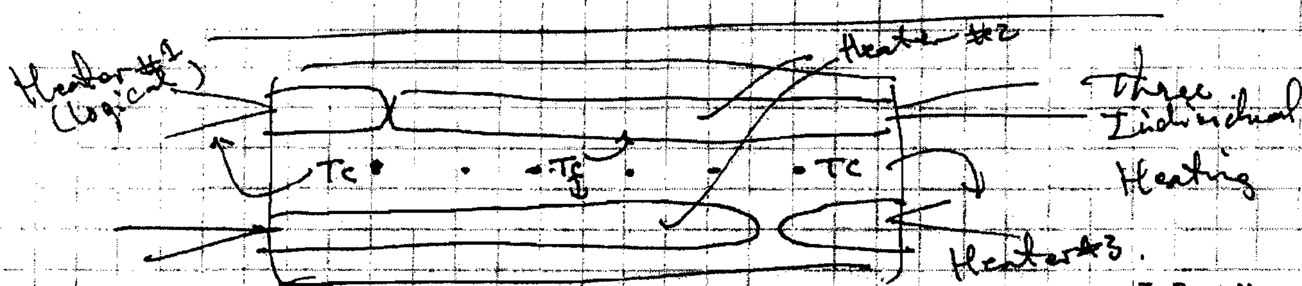
1	2	3	4	5	6
188.3	202.4	201.5	200.3	196.7	188.0

Temperature Setting 300.0°C @ Ch 4 for 1 hour

1	2	3	4	5	6
282.8	302.5	301.5	300.0	295.4	283.8

Dan's Cat 102-11-B / Ch1 flow 18.7% O₂ in N₂ 105ccm Ch2. 25% Ethane 505ccm

		1	2	3	4	5	6	Setting @ Ch 4
Cat loading (mg)		53.4	52.3	56.7	53.8	50.4	52.2	
CX1004	2005							~~200°C~~ 200°C
CX1005	2006	235.6	251.8	251.8	250.0	245.7	236.4	250°C
CX1007	2008	282.9	302.0	301.8	300.2	295.1	284.3	300°C
CX1008	2009	329.7	352.1	351.9	349.8	343.6	330.1	350°C
CX1009	2010	378.7	402.3	402.4	400.4	394.1	379.7	400°C
CX1010	2011	429.5	453.4	453.7	451.8	445.7	430.9	450°C



To Page No.____

Witnessed & Understood by me, RBBonde | Date | Invented by [signature] | Date
Recorded by S. [signature]

TITLE	Project No.	93
	Book No.	

From Page No.

Compensated Heating System control ch for main Heaters

Parallel React
PR_DATA/
12_31_97_1

	1	2	3	4	5	6		
10:01:53	200.2,	203,	200,	199	200	200.2	CX1001	CX2001
10:27:17	249.0	255	251	249	250	249.5	CX1002	CX2002
10:51:09	298.0	305	301	299	299	299.5	CX1003	CX2003
11:31:09	349.9	356	350	348	349	350.0	CX1004	CX2004
11:56:25	395.4	405	400	398	399	399.8	CX1005	CX2005
12:50:36	447.2	455	450	448	449	449.8	CX1006	CX2006
	~~304.0~~	~~344~~	~~350~~	~~357~~	~~367~~	~~376.3~~		
Set	320		340			370		
14:01:36	317.7	336	342	349	359	367.9		07
14:09:44	319.5	336	340	347	358	369.2		08
14:27:21	319.8	336	340	347	359	369.8		09

To Page No.

Witnessed & Understood by me, RB Berude

Date

Invented by [signature]

Recorded by S. Gu

Date

Project No.______
Book No.______
TITLE PR Calibration #2

111

From Page No.___

Matheson Certified Gas STD 6 Component (MR1543)

	Prep.	Actual
CO	2.0%	1.97%
CO_2	2.2%	1.97%
O_2	4.0%	3.89%
Ethylene	6.0%	6.06%
Ethane	30.0%	30.40%
N_2	—	Balance

Calibration gas @ ch 1 @ 30% ~ 3 sccm

11 runs collected @

Data on Enigma / Storage / ParallelReactor / PR_Data
11-2-98-1

CX1006 – CX1010 } used for calibration.
CX2006 – CX2010 }

To Page No.____

Witnessed & Understood by me, RBBonde

Date

Invented by [signature]

Recorded by [signature]

Date